

**Amendments to the Drawings:**

There are no Amendments to Drawings.

## **REMARKS/ARGUMENTS**

### **Summary of Telephone Interview**

The Examiner graciously granted a telephone interview to Inventor Laszlo Keeskes in Aberdeen Proving Ground, APG Maryland and Government Patent Attorney A. David Spevack. The essentials of what was described to the Examiner are incorporated in the remarks below and the attached Declaration.

### **Discussion**

Applicants have noted the Examiners comments regarding the declaration. Applicants contend that the declaration shows a comparison of the best performers within each group and therefore is relevant and dispositive.

The intent of Gu et al. is to systematically study the effect of enriching a Zr-based metallic glass alloy, i.e., Vitreloy 105 (Zr52.5Ti5Cu17.9Ni14.6Al10), with substituting Hf for Zr, demonstrating the extreme difficulty of forming a good glass forming alloy. Although, a reverse dilution of a Hf-based alloy is possible with Zr, the intent of the Applicants, in contrast to that of Gu, is to rely on Gu's data to produce a purely Hf-based metallic glass alloy without the presence of Zr. Vitreloy 105 has an excellent glass forming ability, as demonstrated by a wide undercooled region, low liquidus temperature, high reduced glass transition temperature, and a limited number of crystalline species upon devitrification.

This reference, cited by the examiner, remains at issue. The intent of Gu is to produce a series of higher-density metallic alloys starting with a good glass forming Zr-based alloy, Vitreloy 105 and systematically enriching it with Hf to increase the overall alloy density. Although the density is found to increase, Gu finds that the substitution of Hf leads to unexpected degradation of glass forming ability. The reference makes several key observations:

it is difficult to form a good glass forming alloy based on direct Hf substitution for Zr.

the glass forming ability monotonically decreases, as measured by the reduced glass transition temperature; and

the number of crystallization peaks (species) increase from one to three as the amount of Zr is decremented to 0.

These effects are attributed to the affinity of Zr and Hf for the other alloy components. The simultaneous competition for Cu, Ni, Al, and Ti by both Zr and Hf creates more confusion in the crystallization kinetics, however, not in a beneficial manner. It is further explained that the nucleation kinetics and subsequent growth of species are different in the Zr- and Hf-based systems, respectively. No further explanation is offered in the reference.

The Applicants have been familiar with the work of Gu; see acknowledgements in the reference. They have postulated that the observed differences are attributed to different locations of the respective eutectics, or lowest melting point compounds, for the Zr-Cu-Ni and Hf-Cu-Ni ternary subsets. The eutectic location in the Hf-Cu-Ni system is non-obvious from that of the Zr-Cu-Ni system. Whereas, the Zr-Cu-Ni eutectic is roughly at Zr<sub>67</sub>Cu<sub>17</sub>Ni<sub>16</sub>, the Hf-Cu-Ni 'eutectic' is at Hf<sub>55</sub>Cu<sub>30</sub>Ni<sub>15</sub>. As shown in Figure 1, this is demonstrated from an existing phase assessment for the former and from experimental data generated by the Applicants for the latter.

The intent of the Applicants is to produce a metallic glass alloy based on Hf only. While Claim 1 implies the presence of more than five components, unless the ratio of Cu:Ni is fixed per Claims 7 and 8, similar results are obtained as those of Gu. Adjustment of the value of x to about 0.8 to 0.85 will result in a metallic glass that has poor glass forming ability. This is demonstrated by Applicants' data, shown in Figure 2 and 3, wherein Gu's results are reproduced. The ratios of glass transition temperature to liquidus are lower, but within the same data set, they are consistent with Gu's data. The Trg values cannot be directly related to Gu's, most likely Gu used purer elemental constituent than did the Applicants.

The Applicants demonstrate their effort on improving the glass forming ability of a Hf-based metallic glass. As shown in Table 1, within the Hf-based alloy series, as indicated by Trg, a small deviation from the ideal composition leads to a rapid degradation of glass forming ability. The non-obviousness of the Applicants invention is the single exotherm and narrow solidus to liquidus transition exhibited by the ideal composition, as contrasted to the multiple number of exotherms and wider solidus-to-liquidus transition of the Hf equivalent of Vitreloy 105. See Figures 4 and 5.

Applicants' invention is clearly differentiated from that of Gu and the claims are now in condition for allowance.

This paper is accompanied by a provisional request for a two month extension of time to respond to the Final Rejection and provide time for the Examiner to consider the Amendment and Declaration. Applicant respectfully requests that a timely Notice of Allowance be issued in this case.

The Director is hereby authorized to charge any additional fees or underpayments under 37 C.F.R. § 1.16 & 1.17; and credit any overpayments to Deposit Account No. **19-2201** held in the name of U.S. Army Materiel Command.

Respectfully submitted,  
Intellectual Property Counsel  
U.S. Army Research Laboratory

By \_\_\_\_\_  
A. David Spevack  
Reg. No. 24,743  
Tel.: 301-394-1714  
FAX: 301-394-3972  
e-mail: [dave.spevack@us.army.mil](mailto:dave.spevack@us.army.mil)

ATTN: AMSRD-ARL-0-CC-IP  
2800 Powder Mill Road  
Adelphi, Maryland 20783-1197

4 December 2008

APPENDIX:

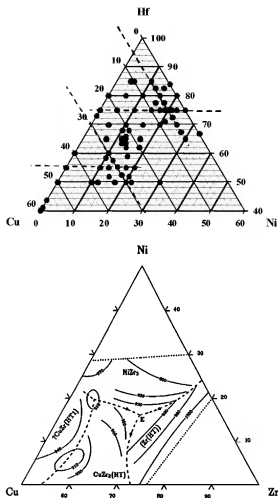


Figure 1. Locus of all experimentally fabricated alloys in Hf-Cu-Ni ternary composition space and the location of the eutectic (E) in the Zr-Cu-Ni system.

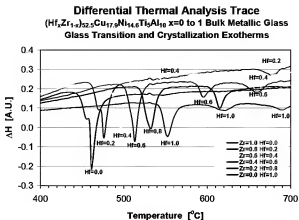


Figure 2. Crystallization events in the Hf-substituted Vitreloy 105 composition series, showing the increasing number of exotherms with decreasing Zr content

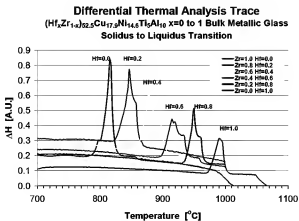


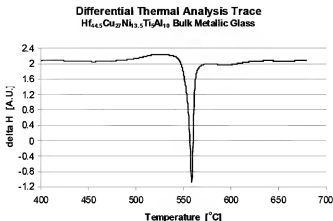
Figure 3. Solidus to liquidus transition in the Hf-substituted Vitreloy 105 composition series, showing the increasing differential melting of multiple crystalline species in each alloy; width of transition

Table 1. Deviations from the ideal composition and their effect on T<sub>g</sub>.

Ingot #	Rod #	Composition (atomic %)	Liquidus (°C)	T <sub>g</sub> onset (°C)	T <sub>g</sub>
20503-6		Hf47 Cu26 Ni13 Al10 Ti5	985		
	020503-6SC			483	0.601
21203		Hf44.5 Cu27 Ni13.5 Al10 Ti5	983		
	21203-4			499	0.615
21903-3		Hf46.75 Cu25.5 Ni12.75 Al10 Ti5	986		
			1024		
	21903-3-2			485	0.602
					0.584
21903-2		Hf44.5 Cu27 Ni13.5 Al10 Ti5	984		
	21903-2-1			494	0.610
42103-1		Hf49 Cu24 Ni12 Al10 Nb5	1042		
	42103-1-1			501	0.589
42103-2		Hf44.5 Cu27 Ni13.5 Al10 Nb5	1036		
	42103-2-1			507	0.596
61003-1		Hf44.5 Cu29 Ni11.5 Al10 Ti5	1024		
	61003-2-1			497	0.594
61003-1		Hf44.5 Cu25 Ni15.5 Al10 Ti5	987		
	61003-1-2			488	0.604
62003-1	62003-1-2	Hf46.5 Cu27 Ni11.5 Al10 Ti5	1005	(observed)	
				481	0.590
62003-2	62003-2-1	Hf42.5 Cu27 Ni15.5 Al10 Ti5	1038		
			1040	(observed)	
				513	0.599
62403-1	62403-1-1	Hf46.5 Cu25 Ni13.5 Al10 Ti5	987		
				501	0.614
				496	0.610
				498	0.612
62403-2	62403-2-1	Hf42.5 Cu29 Ni13.5 Al10 Ti5	1013	(observed)	
				519	0.616
80103-2	80103-2-1	Hf39.5 Ti5 Nb5 Cu27 Ni13.5 Al10			
				No T <sub>g</sub>	



(a)



(b)

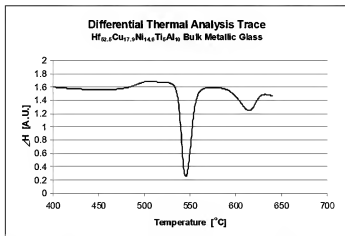
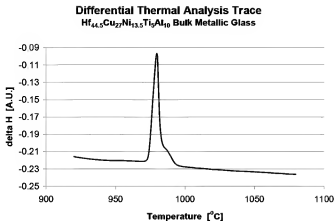


Figure 4. (a) crystallization event of the alloy claimed in the invention, showing a single exotherm. (b) crystallization events of the fully substituted Vitreloy 105 alloy, showing the first two exotherms. The third exotherm was not plotted for convenience.

(a)



(b)

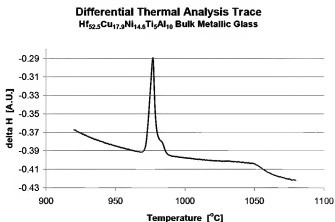


Figure 5 (a) solidus to liquidus transition of the alloy claimed in the invention, showing a narrow endotherm  
(b) solidus to liquidus transition of the fully substituted Vitreloy 105 alloy, showing a significantly wider endotherm